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Nanosecond control and high-density production of spin-polarized hydrogen atoms**

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Abstract

We demonstrate directly the nanosecond-timescale production of spin-polarized hydrogen (SPH) atoms from photodissociation of thermal HBr molecules, and the spin-state and Doppler-resolved detection using polarized fluorescence at 121.6 nm, without requiring hyperfine resolution. These techniques allow a variety of spin-dependent, nanosecond pump-probe experiments with SPH, which were not previously realizable. The possibility of surpassing the SPH densities and production rates of current techniques is discussed.

Main text

Spin-polarized hydrogen (SPH) atoms have wide-ranging uses and potential applications, particularly in the measurement of spin-dependent scattering effects.[1] Examples include studies of the spin-structure of the proton,[2] proposals for the production of spin-polarized anti-protons,[3] uses in nuclear fusion,[4] and the characterization of surface magnetism from surface scattering and chemisorption.[5,6,7] However, despite the fact that hydrogen is the simplest atom and is a natural choice for fundamental studies of spin-dependent collision processes, many such experiments are particularly challenging because of difficulties in both the production and the detection of SPH, especially optically,[8] due to the sub-Doppler spin-orbit splitting of the 2p state at room temperature, and the difficulty in producing intense continuous wave 121.6 nm light for optical pumping of the 2p←1s transition.[9] For collision experiments, conventional methods for SPH production use large and involved experimental setups, such as Stern-Gerlach separation,[10,11] or spin-exchange optical pumping,[1] that achieve densities of only up to about 10^{12} cm^{-3}.[11,12] SPH is usually detected non-remotely with atom polarimeters, which have limited time and spatial resolution, or optically with fluorescence at 121.6 nm, which has been achieved with spin-state selectivity *only with hyperfine resolution*, requiring the SPH translational temperature to be colder than about 80 K.[13]

Recently, a new method for SPH production was demonstrated: the pulsed-laser photodissociation of HCl or HBr at 193 nm, in a supersonically cooled skimmed molecular beam of about 15 K.[14,15] However, the SPH was not detected directly: the degree of polarization of the SPH was inferred from the measurement of the halogen cofragment polarization, which does not allow direct monitoring and use of the SPH.

Here, we extend this method by photodissociating HBr at room temperature and at high pressures [see later], and show that the production of SPH is achieved under these conditions, i.e., unlike many other cases of molecular photodissociation, the photodissociation mechanism of HBr does not change significantly from 15 K to 300 K. We directly detect the SPH on the nanosecond timescale with a

variant [it is a variant, but can we change wording somewhere here to emphasize original/new] of polarized fluorescence at 121.6 nm.[16] The detection scheme allows complete SPH spin-state and Doppler selectivity, without requiring hyperfine resolution and is independent of the SPH translational temperature, while retaining the excellent detection sensitivity of Lyman-α fluorescence.[also want to say polarization sensitivity is high] The combination of these two pulsed-laser techniques, and the characterization of the photodissociation of HBr at room temperature, allows us to infer that pulsed-densities in excess of 10^{17} cm^{-3} are attainable (several orders of magnitude greater than current techniques), and allows the nanosecond-timescale production and detection of SPH in a tabletop setup, which greatly simplifies the study of many spin-dependent collision experiments.

SPH atoms were formed by a 5 ns pulse of circularly polarized ultraviolet (193 nm) light, which was used to photodissociate a room-temperature sample of hydrogen bromide (HBr) gas, at a pressure of about 10 μbar. The density of the SPH produced can be controlled by both the density of the target gas, and the flux of ultraviolet light. The mechanism of SPH production from HCl or HBr photodissociation is described elsewhere.[14,15,16,17]

The SPH detection scheme is experimentally and conceptually straightforward. The H atoms are excited from the 1s $^2S_{1/2}$ to the 2p ^2P_J states using a circularly polarized 121.6 nm light pulse. The 121.6 nm fluorescence of the excited atoms is detected with a photomultiplier tube perpendicular to the propagation of the excitation laser, and a polarizer is used to detect only fluorescence that is linearly polarized along the Z^{lab} axis (see Fig 1a). The geometry of the experiment has been chosen so that angular momentum selection rules constrain the excitation and fluorescence processes to be allowed for only one of the spin-states, so that the H-atom spin-polarization can be determined along the Z^{lab} axis (see Fig. 1b).

Using the Doppler effect, the wavelength of the probe light can be tuned to detect selectively SPH atoms that are moving with velocity projection v_z along the Z^{lab} axis. For the present experiment, the probe-laser bandwidth of about 0.8 cm^{-1} gives a velocity resolution of about 3000 m s^{-1}, but which may be reduced to at least 0.007 cm^{-1} (giving a velocity resolution of about 25 m s^{-1}).[18] The detection scheme time resolution is determined by the pulse widths of the photolysis and probe lasers (each about 10 ns), and by the spin-orbit coupling time in the 2p state of $\tau_{2p} \sim 0.1$ ns. The 2p state can be depolarized by collisions, reducing the sensitivity of the detection scheme to SPH polarization. The present experiment was conducted at pressures of 10 μbar: however, SPH was observed at pressures up to 30 mbar of HCl/He mixtures.

Linearly polarized 121.6 nm light is generated by frequency tripling 364.8 nm light in a krypton/argon gas mixture, which is focused into the vacuum chamber with a magnesium fluoride (MgF_2) lens. The 121.6 nm light is circularly polarized by passing it, under vacuum, through a tilted MgF_2 plate; the tilt

angle (about an axis at 45° to the 121.6 nm linear polarization axis) is varied until the fluorescence intensity along the X^{lab} and Y^{lab} axes is equal, which occurs for circularly polarized light only. The fluorescence is linearly polarized along Z^{lab} by reflecting it from a MgF_2 plate at Brewster's angle.

In Fig. 2a we show the fluorescence signal of SPH from HBr photodissociation using left (L) and right (R) circularly polarized photolysis light, as a function of wavelength of left (L) circularly polarized probe light, to give profiles (LL) and (RL). The alteration between the (L) and (R) photolysis laser polarization states is actuated on a shot-to-shot basis (operating at 10 Hz) using a photoelastic modulator. The visibly clear difference between the two profiles is proportional to the degree of polarization of the SPH. The H atom speeds are about 22.6 km s^{-1} (within a blurring of about 9% mainly due to the spin-orbit splitting of the Br atom cofragment) forming a spherical shell in velocity space (Fig. 2b). The one-dimensional Doppler projection of this spherical shell, along with the polarization effects, gives the experimental signals. The signal combinations I_S=(LL)+(RL) and I_D=(LL)-(RL) are the polarization *independent* (proportional to the velocity distribution only) and the polarization *dependent* signals, respectively, which can be written as:[19,20]

$$I_S = I_0[1-\beta(3\cos^2\theta-1)/4] \quad (2a)$$

$$I_D = I_0[P_Z(1-\beta/2)\cos^2\theta + P_X(1+\beta/4)\sin^2\theta]/2 \quad (2b)$$

where cosθ is the normalized Doppler shift of the SPH atoms, given by the velocity projection $v_z/|v|$ and ranges from -1 to +1, the parameter β describes the spatial distribution of the SPH atom velocities and β=–0.78 for HBr, P_Z is the electron polarization for atoms traveling parallel Z^{lab} (cosθ = 1), and P_X is the electron polarization for atoms traveling perpendicular to Z^{lab} (sinθ = 1). The polarization components P_Z and P_X arise from different photodissociation mechanisms [$P_Z = \sqrt{3}\, a_0^1(\perp)$, and $P_X = \sqrt{3/2}\, \mathrm{Re}[a_1^1(\|,\perp)]/(1+\beta/4)$].[19,20,21] The sum and difference profiles of HBr were analyzed using equations 2a and 2b, and the values of P_Z and P_X are plotted in Fig 2c, along with quantum mechanical ab initio calculated values,[22] and predictions using conservation of angular momentum from previous measurements of the Br cofragments.[15]

The values of P_Z and P_X correspond to the nascent electron polarizations, whereas the proton polarization is initially unpolarized; after 0.7 ns, the polarization is shared between the electron and the proton, and the long-time average polarizations for both the electron and proton are half the values shown in Fig 2c. However, methods have been proposed to polarize the proton before photodissociation, so that the final SPH polarization values need not be reduced,[23] or the electron

polarization can be maintained using a magnetic field greater than about 100 mT,[11] without being depolarized by the proton.

For the HERMES target at DESY (Deutsches Elecktronen-Synchrotron),[24] and the Relativistic Heavy Ion Collider (RHIC) at Brookhaven National Laboratory,[12] the SPH production rates are about 10^{17} atoms s^{-1}. Recently, about 10^{18} atoms s^{-1} has been achieved using spin-exchange optical pumping.[25] The SPH production rate from molecular photodissociation is limited by the photodissociation photon flux; existing commercial pulsed excimer lasers can produce about 10^{20} photons s^{-1} at 193 nm. If necessary, the fast-moving SPH can be separated from the slow halogen atoms by photodissociating from a molecular beam, and by detecting the SPH at some distance.

The combination, in a table-top setup, of the pulsed-laser production of SPH with the pulsed-laser detection of SPH described here, will allow experiments not previously possible, such as remote detection SPH at low density under single collision conditions at nanosecond pump-probe delays. This includes experiments such as the scattering of SPH from surfaces for the investigation of surface magnetism,[7] and the study of the molecular photodissociation mechanisms of important H-containing molecules (e.g. H_2, H_2O, CH_4 and other hydrocarbons), which produce SPH via both coherent and incoherent photodissociation mechanisms.[19-22]

Figures

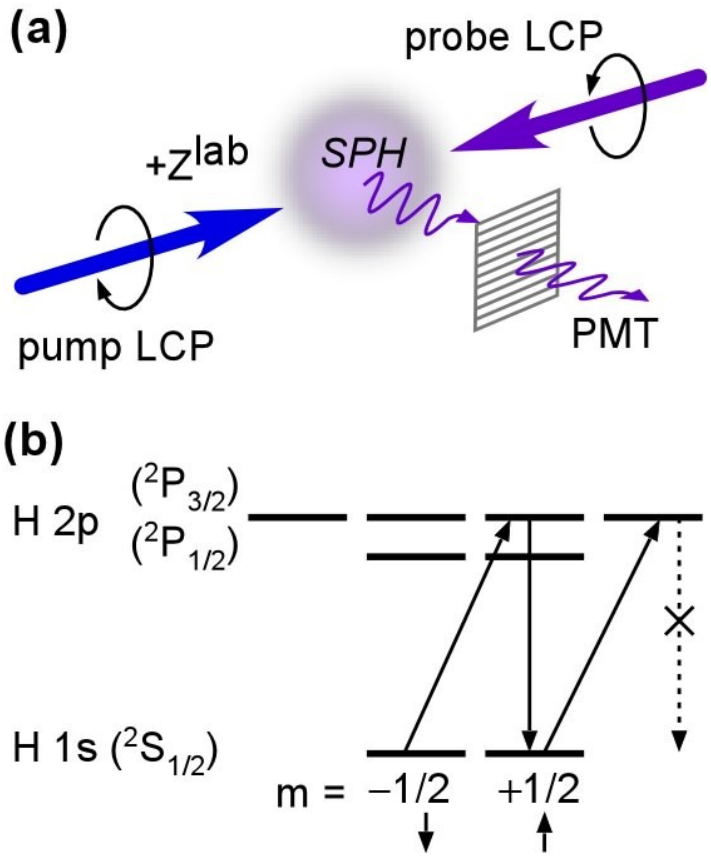

Figure 1. SPH detection scheme. (a) Schematic diagram showing the laboratory $+Z^{lab}$ axis, defined by the direction of the left circularly polarized (LCP) 193 nm pump light pulse that dissociates HBr to produce SPH, the counterpropagating probe laser, and the photomultiplier tube (PMT) collecting fluorescence perpendicular to Z^{lab}, but linearly polarized parallel to Z^{lab} . (b) Energy-level detection scheme, showing that only the m = -1/2 spin state is detected using LCP 121.6 nm probe light.

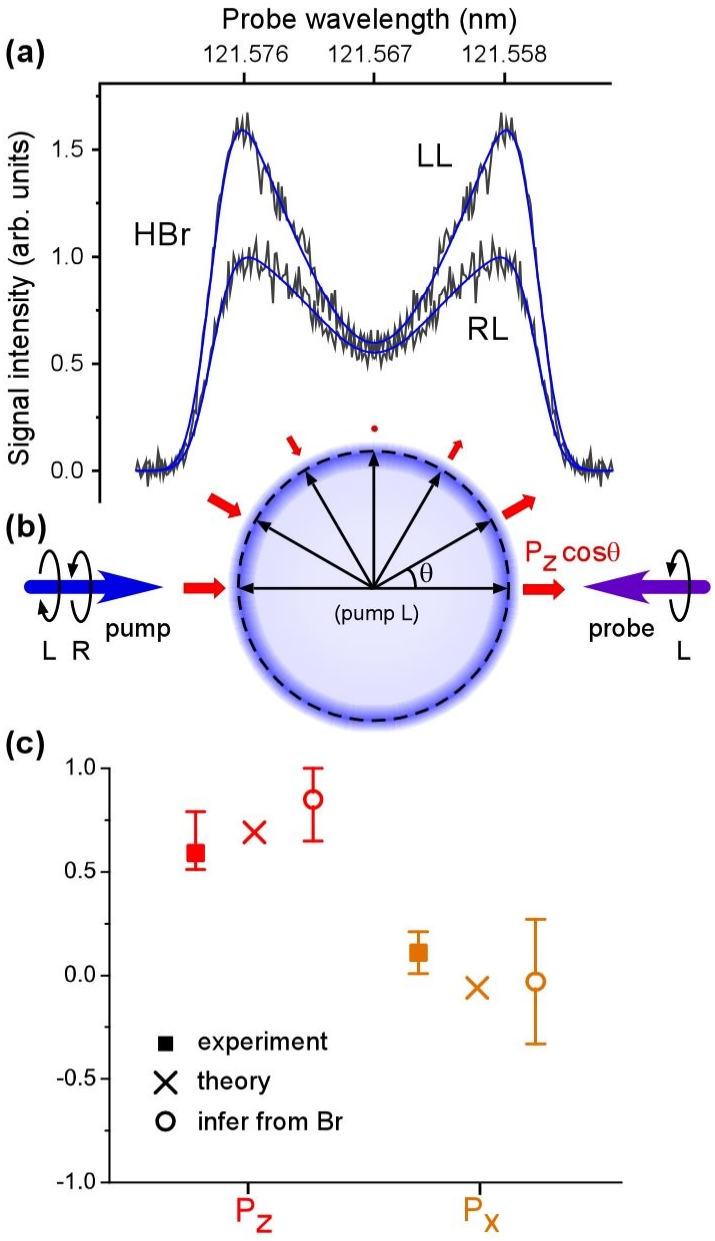

Figure 2. SPH detection from HBr photodissociation at 193 nm. Experimental fluorescence signals as a function of wavelength of the left (L) circularly polarized probe light. (a) Traces taken using alternately left (L) and right (R) circularly polarized photolysis light. (b) The spherical velocity distribution of the SPH, showing the SPH polarization (red arrows) as a function of angle with respect to the laser propagation axis. The one-dimensional projection of this distribution gives the experimental signals. (c) Analysis of the experimental profiles, using Equation 2, yields values of P_Z and P_X for HBr shown here (solid squares), and are within error of ab initio calculated values (crosses), and values inferred from the Br cofragment polarizations (open circles). The lower error bars represent 2σ of the fitted values. The upper error bars also include the uncertainty in the degree of circular polarization of the 121.6 nm light.

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